UCRL-JRNL-214579

LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

Cost-Reducing Methods for Thin Film Solid Oxide Fuel Cells

R. S. Glass

August 12, 2005

Materials Research Society Bulletin

Disclaimer

Cost-Reducing Methods for Thin Film Solid Oxide Fuel Cells

Lawrence Livermore National Laboratory (LLNL) has developed an improved method for fabrication of a thin film Solid Oxide Fuel Cell (SOFC) using a colloidal technique. Dense, crack-free, yttria-stabilized-zirconia (YSZ) films of up to 100 microns thick were deposited on nickel oxide/YSZ substrates and porous $La_{0.85}Sr_{0.15}MnO_3$ (LSM) substrates. The new technique was also used to deposit a compositionally-graded film of YSZ and $Ce_{0.8}Y_{0.2}O_2$ (CYO), which is useful for matching the thermal expansion coefficient to an adjacent layer.

The SOFC is a solid-state electrochemical device that converts the chemical energy of fuel directly into electricity. Its efficiency and low emission positions SOFC on the short list of power generation technologies for the 21st century. However, commercialization efforts are hampered in part by the high cost of fabrication. One area of high cost is the fabrication of thin, defect free coatings. It is desirable to produce coatings that are in the range of 10-40 microns to minimize resistance yet maintain mechanical integrity.

Deposition of films thicker than 10 microns in a single step using conventional techniques such as dip coating, spin coating, slurry painting or electrophoretic deposition generally results in cracking due to shrinkage when the solvent volatilizes. Therefore, generation of thicker coatings requires repeated thin film deposition, leading to long processing times and higher cost. LLNL's Colloidal Spray Deposition (CSD) method evaporates the solvent upon contact with the substrate, depositing a dense, finely divided powder film. This is achieved by heating the substrate to a temperature above or close to the boiling point of the solvent. When the fine mist is sprayed onto the hot substrate, the solvent evaporates rapidly, leaving a compact layer of powder. Continuous removal of solvent during extended deposition results in a thick defect-free film which is subsequently sintered. LLNL has optimized the aspects of technique including the nebulization and spraying process as well as the starting solution compositions. Both dense and porous coatings can be created, resulting in a fabrication method suitable for preparation of both the electrolyte as well as the electrodes.

The CSD technique can be used to simplify fabrication and design of the fuel cell electrolyte and the electrodes. Fig. 1a shows a SEM micrograph of the cross section of a dense, crack-free, porous 13 µm YSZ thin film on a porous Ni/YSZ (anode) disk. Fig. 1b shows a SEM micrograph of the cross section of a 70 µm porous Ni/YSZ film on a YSZ disk. Crack-free films as thick as 100 µm have been deposited.

Fuel cells with CSD-deposited YSZ film on NiO/YSZ substrate and a Pt cathode were tested at 900°C in air/hydrogen where the NiO electrode was reduced to Ni. The open-circuit voltage was 1.1 V at 900°C, close to the theoretical voltage, indicating no leakage in the film. The short-circuit current density was 1.2 $A.cm^{-2}$ and the power density reached 0.55 $W.cm^{-2}$.

The CSD technique can also facilitate the use of materials that can improve fuel cell performance. For example, doped-ceria (CYO) has non-negligible electronic conduction in fuel conditions and cannot serve as an electrolyte by itself. A bi-layer of YSZ and doped-ceria has been proposed where the YSZ layer serves to block electrons. However, cracking and delamination have been observed due to the higher thermal expansion coefficient of doped-ceria. Relaxing the mechanical stress by grading the interface from YSZ to CYO is difficult to achieve using existing thin film deposition techniques. However, LLNL has been able to grade composition of a thin film over several microns (fig.2a). No visible interface was observed. Since YSZ and CYO form a complete solid solution at temperatures higher than 1300°C the graded film is believed to be a single phase material with composition changing progressively from pure YSZ to pure CYO. Fig. 2b shows the concentration profile of the graded film as determined by electron microprobe analysis.

In addition to fuel cells, the CSD technique can be used to fabricate sensors, membranes, and other components. It can also be used to generate chemically inert, durable ceramic or metal coatings for components having different geometries for use in a variety of applications, particularly in harsh environments.

LLNL is seeking industrial partners with a demonstrated ability to bring this technology to the market for collaborative research and development and /or to license the CSD technique, which has been shown to be versatile and economical.
Source: Dr. Annemarie Meike, Industrial Partnerships and Commercialization Office, Lawrence Livermore National Laboratory, L-795, P.O. Box 808, Livermore, CA 94550. Phone: 925 422 3735, facsimile: 925 423 8988, e-mail: meike1@llnl.gov.

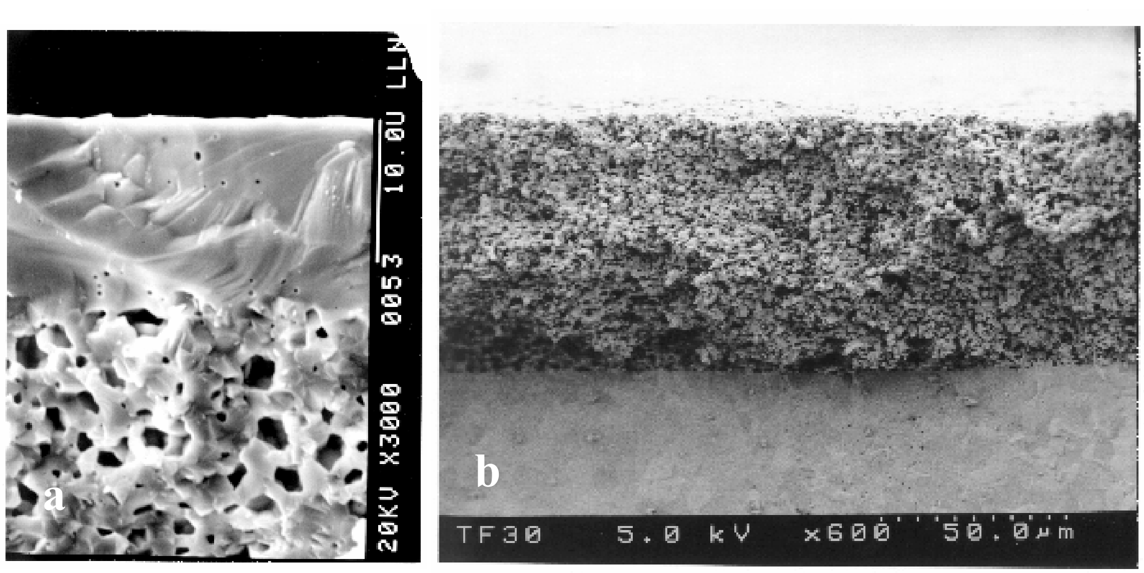

Fig. 1. a) 13 µm YSZ film on Ni/YSZ substrate, and b) 70 µm porous Ni/YSZ film on YSZ disk.

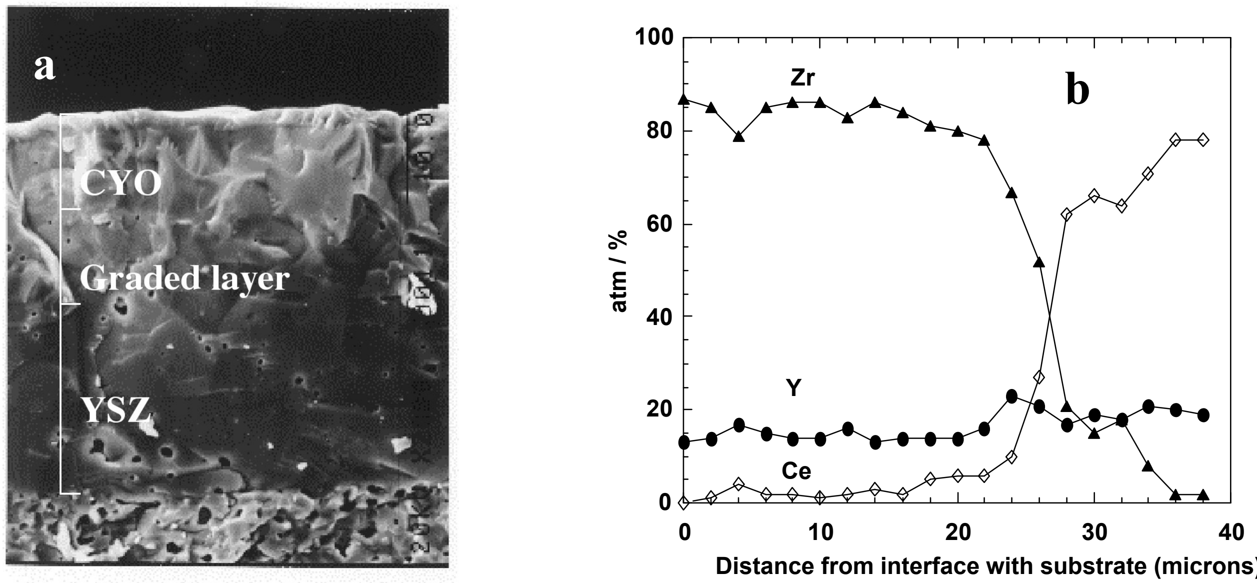

Fig. 2. a) 40 μm YSZ and CYO film with a graded interface, and b) Composition profile of the YSZ/CYO graded film.

This work was performed under the auspices of the U.S. Department of Energy by University of California, Lawrence Livermore National Laboratory under contract W-7405-Eng-48.